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AUTHOR(S) Naomi M. Becker (EM-8)

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Los Alamos Los Alamos National Laboratory
Los Alamos, New Mexico 87545

QUANTIFICATION OF URANIUM TRANSPORT AWAY FROM FIRING SITES AT LOS ALAMOS NATIONAL LABORATORY-A MASS BALANCE APPROACH

by: Naomi M. Becker
Environmental Protection Group (EM-8)
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

ABSTRACT

Investigations were conducted at Los Alamos National Laboratory to quantify the extent of migration of depleted uranium away from firing sites. Extensive sampling of air particles, soil, sediment, and water was conducted to establish the magnitude of uranium contamination throughout one watershed. The uranium source term was estimated, and mass balance calculations were performed to compare the percentage of migrated uranium with original expenditures. Mass balance calculations can be powerful in identification of the extent of waste migration and used as an aid in planning future waste investigations.

DESCRIPTION OF THE PROBLEM

Los Alamos National Laboratory routinely collects and analyzes water, soil, sediment, particulate materials, vegetation, and biota for chemical and radiochemical constituents to assess the Laboratory's impact on the environment. During 1983, fish collected from a reservoir downstream from the Laboratory exhibited elevated levels of uranium that were statistically significant. Investigations were initiated to determine if this elevated uranium could be due to offsite transport of uranium which is used in dynamic weapons testing at Laboratory firing sites and to quantify the extent of migration within the watershed.

During a dynamic weapons test, depleted uranium is substituted for enriched uranium in a weapons component. The component is then explosively detonated, or is impacted against a target in the open air environment. This results in both the production of a wide range of depleted uranium particles as well as particle scattering over a large distance away from the firing pad. The explosive detonation process of aerial distribution over the watershed distinguishes this contaminant transport problems from others where the source term is spatially discrete (e.g., transport away from a waste pile or landfill.)

Mass balance calculations can aid waste management investigations which characterize the extent and magnitude of waste migration. At Los Alamos, applied mass balance to the determination of the extent of uranium transport away from firing sites will be described and will demonstrate how useful a tool this can be in decision-making for waste treatment and cleanup procedures.

SETTING AND SOURCE TERM DESCRIPTION

Although there are numerous watersheds at the Laboratory which contain firing sites where dynamic tests are conducted, investigations were confined to one watershed named Potrillo Canyon. Potrillo Canyon was selected because of its small size, it is contained entirely within the Laboratory boundaries, it is limited to public access, and contains five firing sites, four of which remain active today. Potrillo Canyon is about 7.8 km^2 in area, 8 km in length, and is relatively steep, with an average gradient of 3 percent. The watershed is characterized by flat mesa tops leading to nearly vertical canyon walls which terminate in large talus piles of boulders of Bandelier Tuff, a volcanic rock composed of ash flows and ash falls.

In terms of historical usage of uranium, it has been estimated that between 85,000 and 105,000 kg of depleted and natural uranium has been expended by Los Alamos National Laboratory since the 1940's. Uranium usage was greatest during the early years of Laboratory operation; it is assumed that as much as 45,000 kg of (depleted) uranium was used between 1943 and 1953. A conservative estimate of the total uranium source term in Potrillo Canyon is about 35,000 kg (1).

RESULTS OF DEPLETED URANIUM SAMPLING IN SOIL, SEDIMENT, AIR, AND WATER

More than 450 samples of fallout from air, soil, sediment, and water and suspended sediment in spring/summer/autumn runoff were collected between 1983 and 1990 and analyzed for total uranium to evaluate the magnitude of transport of uranium away from firing sites by airborne and surface water runoff mechanisms. Results for the maximum, minimum and mean values are presented in Table I. Background levels of uranium in fallout range from 1-6 μg/g, in soil from 2-5 μg/g, and in water about 1 ppb (1). The greatest concentrations of uranium were found in transported suspended sediment carried in runoff waters where average concentrations were 51.1 μg/g, followed by sediment present in stream banks where average concentrations were 42.2 μg/g, Table I. Average concentrations of 17.5 μg/g were observed in geomorphologic deposits such as alluvial fans and point bars. Average uranium concentrations dissolved in runoff waters of 11.9 ppb were also found to be elevated above background concentrations. Uranium present in fallout and in surface soils were found to be at or slightly above background concentrations in most samples, which indicated that airborne transport and wind redistribution is not significant in mobilization of uranium away from firing sites. Uranium concentrations in runoff in the dissolved and suspended sediment phases were found to decline with downstream direction in the watershed, with the largest concentrations below two firing sites near the top of the watershed, implying both dilution and contaminant deposition in the distal direction.

MASS BALANCE CALCULATIONS

Calculations were made to determine the amount of uranium currently coexisting on or attached to fluvial sediment in the watershed today. Using average measured concentrations of uranium in fluvial sediment and subtracting off background levels of uranium, estimates were made of the uranium inventory in the channel, on banks, in point bars and alluvial fans, and in an area known as a discharge sink where sediment is

preferentially accumulating in the watershed. Calculations were made considering uranium concentrations above background of: (1) 3 ppm (by weight) along the entire channel length and width to a depth of 0.1 m in the channel bed; (2) 3.5 ppm above background along the entire channel length on both banks extending 1 m from the bank edge and 0.1 m depth; (3) 7 ppm in an estimated 30 point bars deposits upstream from the discharge sink; (4) 9 ppm in 2 major alluvial fans; and (5) 1 ppm above background in a 0.2 m depth profile within the discharge sink. For each of these 5 regions, soil masses were multiplied by soil concentrations to obtain uranium volumes. For the channel and bank segments, point bar deposits and major alluvial fans upstream of the discharge sink, it was estimated that between 100 and 300 kg of uranium are present. This quantity represents less than 1 percent of the estimated total uranium expenditure (35,000 kg).

From these data it may be concluded that most of the uranium mass 1) is not tied up in the fluvial sediments, 2) has already left the watershed or 3) remains on the firing sites. Flow and uranium loss can occur by vertical flow (infiltration) in the discharge sink or through horizontal flow out the watershed. Infiltration and surface water losses are considered separately.

Examining the volume of uranium which enters the discharge sink, there are dissolved and suspended uranium components. Assuming an annual total inflow of 5200 m^3 (measured during 1990) and an average dissolved uranium concentration of 1.86 ppb (measured between 1984 and 1990), then 9.5 g of uranium annually are carried in the dissolved phase. Over 45 years of operation this would amount to an influx of about 0.5 kg of dissolved uranium transported into the discharge sink, or less than 1 percent of the estimated 35,000 kg source term.

The average annual suspended sediment load was calculated by assuming the suspended load to be 5 percent of the average discharge based upon visual observations of the volume of suspended sediment which was collected in cumulative samplers emplaced throughout the watershed. Using a range of 35,000 to 1,400,000 kg/km^2/yr (3) and multiplying by an average suspended sediment uranium concentrations of 8.01 ppm by weight (measured), the average annual uranium influx into the discharge sink ranged from 1 to 36.5 kg/yr. The combined dissolved and suspended sediment influx to the discharge sink over the 45 years constituted between 0.1 and 4.7 percent of the 35,000 kg uranium source term.

If large volumes of depleted uranium had exited the watershed through surface water transport at the outlet, a depleted uranium signature observable through inspection of the ratio of uranium-235 to uranium 238 is expected to have remained in the sediments in the lower half of the watershed. Because little depleted uranium signature was observed in sediments in the channel, banks, and floodplain downstream of the discharge sink, and it was inferred through chemical and aerial photographic data that there has been little transport across the discharge sink during the last 23 years, it was assumed that most of the uranium must remain in the watershed.

A second calculation was made to determine what the concentrations of uranium in runoff water should be if all the uranium expended were uniformly dissolved in precipitation on an annual basis. Considering 0.5 m of precipitation annually and that 80 percent of the precipitation is lost to evaporation, transpiration and infiltration, then,

Dissolved Concentration

$$= 35{,}000 \text{ kg} / (0.2)(0.5 \text{ m})(7.8 \text{ km}^2)(45 \text{ yrs})$$
$$= 1 \text{ ppm.} \qquad \text{(Eq. 1)}$$

A dissolved concentration of one ppm is an underestimate because not all precipitation contacts the uranium; expected concentrations would be even higher. The dissolved concentration of 1 ppm exceeds observed dissolved uranium concentrations in runoff water by 2 to 3 orders of magnitude. Clearly, high dissolved uranium concentrations in surface water are not observed and dissolved transport in surface water is not a main uranium transport mechanism.

The argument that most of the uranium mass has left the watershed either by movement into the discharge sink (dissolved phase) or by flowing past the watershed outlet is rejected. Calculations showed that the fluvial sediment contain less than 5 percent of the expended mass. The only plausible location for the remaining uranium is at or near the firing sites.

Results from an aerial radiological flyover in 1982 (4) estimated that between 4 and 23 Curies of Protactinium-234m (Pa-234m) remained near three firing sites in the watershed, the variability dependent on the estimated vertical distribution. It is reasonable to assume equilibrium between Pa-234m and uranium-238 (U-238) because the half-life decay from uranium-238 to Protactinium is short, on the order of about a half year, whereas the half-life of uranium-238 is long, on the order of 4.5×10^9 years. Then assuming this equilibrium (equality between Pa-234m and U-238), an estimated 4-23 Curies of uranium remain at the three firing sites. Multiplying Curies by 3.003×10^6 to convert to kilograms, the amount of uranium still remaining at the firing sites is calculated to range from 12,000 to 69,000 kg, bracketing the estimated 35,000 kg uranium expended in Potrillo Canyon.

Consider this hypothesis from another viewpoint. If all the 35,000 kg of uranium were situated at the three firing sites, then what magnitude of soil concentration would be expected? Assuming the contaminated area is 26,000 m^3 from measurements with an assumed uniform concentration to 0.6 m depth,

Soil Concentration

$$= 35{,}000 \text{ kg} / (26{,}000 \text{ m}^3 \times 19 \text{ g/cm}^3)$$
$$= 72 \text{ ppm,} \qquad \text{(Eq. 2)}$$

and 19 g/cm^3 is the approximate specific weight of uranium. Unpublished surface soil studies reported concentrations of uranium ranging from 408 to 3359 ppm by weight at one of these firing sites, and unpublished surface and depth data at another of the firing sites ranged from 560 to 4580 ppm uranium by weight. Concentrations in the vertical direction ranged from 2 to 75 ppm by weight to 3.7 m depth with the largest concentrations in the uppermost 0.6 m. Therefore, an average soil concentration of 72 ppm is consistent with measured concentrations at firing sites. This shows that the original estimated source term of 35,000 kg may even be slightly low.

APPLICATIONS TO WASTE MANAGEMENT

In investigations of former waste disposals sites, a frequent objective is to determine the extent of waste migration from its original location. Waste inventories or inventory estimates provide the initial source term. Sampling in the vicinity of the disposal unit can be designed to provide an estimate of the extent of the waste migration. Pathways which might be considered significant could include 1) air, in particulate, gaseous and vapor phases; 2) soil and sediment, with transport by hydrologic mechanisms in both the horizontal (surface water) and vertical (saturated and unsaturated, potentially multi-phase flow); 3) water transport, by runoff and snowmelt, through infiltration, in the dissolved and suspended sediment phases. Results from sampling are then integrated over the sampling area and compared to the original source term estimates. When the percent of waste which has migrated is small is compared to the original amount, then decisions can be made regarding the need and extent for future sampling, remediation, capping, or possibly exhumation. Risk assessment can be performed as an aid in the decision-making process. In some cases, the combination of inventory analyses and prelimary sampling investigations coupled with mass balance calculations and risk assessment may obviate extensive and costly waste site studies.

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Table I.
Uranium in Air, Water, Sediment, and Soil
Units are μg/g (except where noted)

	Min	Max	Mean	Standard Deviation
Air (fallout)	0.8	7.5	3.5	2.1
Soil (top 5 cm)	1.2	66.	4.8	8.3
Runoff				
- dissolved (ppb)	BDL*	654	11.9+	53.4+
- suspended sediment	0.5	404.9	51.1	157.1
Sediment				
- Channel Deposits	1.0	158.1	8.6	23.0
- Bank Deposits	1.5	373.0	42.2	100.3
- Alluvial Fans and Point Bars	1.6	154.5	17.5	39.8

*Below Limits of Detection.
+Derived using Maximum Likelihood Estimators (3).